

# Pure superposition states of atoms generated by a bichromatic elliptically polarized field

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We find specific polarizations of components of a bichromatic field, which allow one to prepare pure superposition states of atoms, using the coherent population trapping effect. These  $m-m$  states are prepared in the system of Zeeman substates of the ground-state hyperfine levels with arbitrary angular momenta  $F_1$  and  $F_2$ . It is established that, in general case  $m \neq 0$ , the use of waves with elliptical polarizations ( $\varepsilon_1 \perp \varepsilon_2$  field configuration for alkali metal atoms) is necessary for the pure state preparation. We analytically show an unique advantage of the D1 line of alkali metal atoms, which consists in the possibility to generate pure  $m-m$  states even in the absence of spectral resolution of the excited-state hyperfine levels, contrary to the D2 line.

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## I. INTRODUCTION

For many applications such as high-precision spectroscopy, frequency standards, magnetometry, lasers, masers, quantum informatics etc. it is important to have a possibility to prepare (in a controllable way) pure quantum states of atoms, which are superpositions of states belonging to different energy levels. The coherent population trapping (CPT) effect [1, 2] is one of possible ways for the superposition state preparation by optical means. In this case the pure quantum state is a dark (non-absorbing) state. For a model three-state  $\Lambda$  system the CPT effect has been well studied [2, 3]. However, for real atoms with complicated Zeeman and hyperfine structures of energy levels, a nontrivial problem on the choice of excitation scheme suitable for the pure superposition state generation is actual [4, 5, 6, 7, 8].

It is worth to formulate two main conditions for the optimal dark resonance detection, when the resonance amplitude and contrast are maximal. i) At the exact two-photon resonance the pure dark superposition state has to exist. ii) Trap dark states insensitive to the Raman detuning should be absent. In opposite case part of atoms will be accumulated in such states in the course of optical pumping, and they will not contribute to the two-photon resonance formation.

In the present paper we theoretically investigate the possibility of the dark superposition state preparation in the system of Zeeman substates of the ground-state hyperfine levels with the total angular momenta  $F_1$  and  $F_2$ . It is assumed that a bichromatic field excites the two-photon resonance of  $\Lambda$  type. General conditions for the polarizations of the frequency components are formulated in the case when the pure dark state is a superposition of the Zeeman substates  $|F_1, m\rangle$  and  $|F_2, m\rangle$  with the same magnetic quantum number  $m$ , i.e. at the condition of the  $m-m$  two-photon resonance. As is well-known, such resonances are used in atomic clocks and magnetometers based on CPT [9, 10, 11]. The main attention is given

to the case of alkali metal atoms, when the angular momenta  $F_1$  and  $F_2$  differ by one. We show that in this case the  $m-m$  dark states are generated by the  $\varepsilon_1 \perp \varepsilon_2$  configuration of the bichromatic running wave, where the major semiaxes of the polarization ellipses are orthogonal. The ellipticity parameters  $\varepsilon_1$  and  $\varepsilon_2$  are coupled by a relationship dependent on the quantum numbers  $m$  and  $F$ . In the particular case of  $m = 0$  the pure dark 0-0 states are prepared in the  $\varepsilon \perp \varepsilon$  field configurations, where the frequency components have the same ellipticity. We consider also the situation when the excited-state hyperfine structure is not spectrally resolved (e.g. due to the optical transition broadening by collisions with a buffer gas). It turns out that for the D1 line of alkali metal atoms all the dark  $m-m$  states are conserved, while for the D2 line such states are absent.

## II. STATEMENT OF THE PROBLEM

We consider the resonant interaction of atoms with a two-frequency field formed by copropagating waves running along  $z$  axis:

$$\mathbf{E}(z, t) = E_1(z)\mathbf{a}_1 e^{-i\omega_1 t} + E_2(z)\mathbf{a}_2 e^{-i\omega_2 t} + c.c. \quad (1)$$

The frequency components of this field have arbitrary complex amplitudes  $E_{1,2}$  and elliptical polarizations described by unit vectors  $\mathbf{a}_{1,2}$ . These vectors can be written in the spherical basis ( $\mathbf{e}_{\pm 1} = \mp(\mathbf{e}_x \pm i\mathbf{e}_y)/\sqrt{2}$ ) as

$$\begin{aligned} \mathbf{a}_1 &= \sin(\varepsilon_1 - \pi/4)\mathbf{e}_{-1} + \cos(\varepsilon_1 - \pi/4)\mathbf{e}_{+1} \\ \mathbf{a}_2 &= e^{-i\theta} \sin(\varepsilon_2 - \pi/4)\mathbf{e}_{-1} + e^{i\theta} \cos(\varepsilon_2 - \pi/4)\mathbf{e}_{+1} \\ -\pi/4 &\leq \varepsilon_{1,2} \leq \pi/4, \end{aligned} \quad (2)$$

where  $\varepsilon_j$  is the ellipticity parameter (angle) of the  $j$ -th component,  $|\tan \varepsilon|$  is equal to the ratio of the minor semiaxes of the ellipse to the major one (see in Fig.1a), and the sign of  $\varepsilon$  governs the direction of the field vector rotation;  $\theta$  is the angle between the major semiaxes of

the polarization ellipses. It is assumed that atoms are being in a static magnetic field  $\mathbf{B}$  directed along the  $z$  axis (see in Fig.1a).

The field (1) drives the two-photon resonance of  $\Lambda$  type between the two ground-state hyperfine levels with the total angular momenta  $F_1$  and  $F_2$  (it does not matter integer or half-integer). The wavefunctions of the Zeeman substates will be denoted as  $|F_1, m\rangle$  and  $|F_2, m\rangle$ . In the case of spectral resolution of the excited-state hyperfine structure the two-photon resonance can be excited (by a suitable choice of the one-photon detuning) via an isolated excited-state hyperfine level with the total angular momentum  $F_e$  (see in Fig.1a), with  $\{|F_e, \mu\rangle\}$  the corresponding Zeeman substate wavefunctions. In what follows we shall use the Dirac representation:

$$e^{-i\mathcal{E}_{F_m}t/\hbar}|F, m\rangle \rightarrow |F, m\rangle,$$

where  $\mathcal{E}_{F_m}$  is the energy of the state  $|F, m\rangle$  with account for the Zeeman shift induced by the field  $\mathbf{B}$ . The dipole interaction Hamiltonian  $-(\hat{\mathbf{d}}\mathbf{E}) = \hat{V} + \hat{V}^\dagger$  in the rotating wave approximation has the form:

$$\begin{aligned} \hat{V} = & \\ d_{F_1 F_e} E_1 \sum_{\mu m q} e^{-i\delta_{\mu m}^{(1)} t} C_{F_1 m, 1 q}^{F_e \mu} (\mathbf{a}_1)^q |F_e, \mu\rangle \langle F_1, m| + \\ d_{F_2 F_e} E_2 \sum_{\mu m q} e^{-i\delta_{\mu m}^{(2)} t} C_{F_2 m, 1 q}^{F_e \mu} (\mathbf{a}_2)^q |F_e, \mu\rangle \langle F_2, m|. \end{aligned} \quad (3)$$

Here  $d_{F_1 F_e}$  and  $d_{F_2 F_e}$  are the reduced dipole matrix elements of the corresponding optical transitions  $F_1 \rightarrow F_e$  and  $F_2 \rightarrow F_e$  (see in Fig.1a);  $C_{F_j m_j, 1 q}^{F_e \mu}$  denotes the Clebsch-Gordan coefficient;  $(\mathbf{a}_j)^q$  are the contravariant spherical components of the polarization vector  $\mathbf{a}_j$  (see eq.(2)); and  $\delta_{\mu m}^{(j)} = \omega_j - (\mathcal{E}_{F_e \mu} - \mathcal{E}_{F_j m})/\hbar$  (where  $j = 1, 2$ ) are the one-photon detunings.

The main objective of the present paper is to determine conditions of the existence of dark states  $|dark^{(m)}\rangle$ , which are a coherent superposition of the ground-state Zeeman sublevels  $|F_1, m\rangle$  and  $|F_2, m\rangle$  with the same angular momentum projection  $m$  with respect to the  $z$  axis:

$$|dark^{(m)}\rangle = A_1 |F_1, m\rangle + A_2 |F_2, m\rangle. \quad (4)$$

These dark states nullify the interaction Hamiltonian (3):

$$\hat{V} |dark^{(m)}\rangle = 0 \quad (5)$$

at the exact two-photon  $m-m$  resonance, when  $\omega_1 - \omega_2 = (\mathcal{E}_{F_2 m} - \mathcal{E}_{F_1 m})/\hbar$ . In the course of the optical pumping process atoms are accumulated in the dark  $m-m$  state, i.e. the pure superposition state (4,5) will be generated.

For the sake of simplicity we shall assume that the ground-state Zeeman splitting exceeds significantly the width of the two-photon  $m-m$  resonances, making them spectrally resolved. As is seen from Fig.1b, the transitions between the states  $|F_2, m\rangle$  and  $|F_1, m\rangle$ , induced by

the  $\sigma_+$  or  $\sigma_-$  circularly polarized field components, form the simple three-state  $\Lambda$  systems. The dark states for each of those  $\Lambda$  systems can be written in the form:

$$|dark_+^{(m)}\rangle = N_{(+)} \times \quad (6)$$

$$\left[ |F_1, m\rangle - \frac{e^{-i\theta} E_1 d_{F_1 F_e} C_{F_1 m, 11}^{F_e m+1} \cos(\varepsilon_1 - \pi/4)}{E_2 d_{F_2 F_e} C_{F_2 m, 11}^{F_e m+1} \cos(\varepsilon_2 - \pi/4)} |F_2, m\rangle \right]$$

$$|dark_-^{(m)}\rangle = N_{(-)} \times \quad (7)$$

$$\left[ |F_1, m\rangle - \frac{e^{i\theta} E_1 d_{F_1 F_e} C_{F_1 m, 1-1}^{F_e m-1} \sin(\varepsilon_1 - \pi/4)}{E_2 d_{F_2 F_e} C_{F_2 m, 1-1}^{F_e m-1} \sin(\varepsilon_2 - \pi/4)} |F_2, m\rangle \right],$$

where  $N_{(\pm)}$  are the normalization constants. Obviously, the dark state  $|dark^{(m)}\rangle$ , nullifying simultaneously the interactions with the  $\sigma_+$  and  $\sigma_-$  polarization components, is in existence if and only if  $|dark_+^{(m)}\rangle = |dark_-^{(m)}\rangle = |dark^{(m)}\rangle$ . This condition is satisfied, when:

$$\begin{aligned} \frac{e^{-i\theta} C_{F_1 m, 11}^{F_e m+1} \cos(\varepsilon_1 - \pi/4)}{C_{F_2 m, 11}^{F_e m+1} \cos(\varepsilon_2 - \pi/4)} = \\ = \frac{e^{i\theta} C_{F_1 m, 1-1}^{F_e m-1} \sin(\varepsilon_1 - \pi/4)}{C_{F_2 m, 1-1}^{F_e m-1} \sin(\varepsilon_2 - \pi/4)}. \end{aligned} \quad (8)$$

It is more convenient to write down this equation as

$$\frac{C_{F_2 m, 11}^{F_e m+1} C_{F_1 m, 1-1}^{F_e m-1}}{C_{F_1 m, 11}^{F_e m+1} C_{F_2 m, 1-1}^{F_e m-1}} e^{i2\theta} = \frac{\tan(\varepsilon_1 + \pi/4)}{\tan(\varepsilon_2 + \pi/4)}. \quad (9)$$

Now we are ready to analyze in detail different variants of the total angular momentum values  $F_1$  and  $F_2$ .

### III. ALKALI METAL ATOMS

Consider the case, when  $F_1$  and  $F_2$  differ by one, as it takes place, for example, for alkali metal atoms. For definiteness, let  $F_1 = F$  and  $F_2 = F + 1$ . Owing to the selection rules for dipole transitions the excited-state angular momentum  $F_e$  can acquire just two values  $F$  and  $F + 1$ .

Using explicit algebraic formula for the Clebsch-Gordan coefficients, we find that the ratio in the l.h.s. of eq.(9):

$$\frac{C_{(F+1)m, 11}^{F_e m+1} C_{Fm, 1-1}^{F_e m-1}}{C_{Fm, 11}^{F_e m+1} C_{(F+1)m, 1-1}^{F_e m-1}} = -\frac{1 + F - m}{1 + F + m} \quad (10)$$

does not depend on the value  $F_e = F, F + 1$ , and it is negative. Therefore, taking into account the positivity of the r.h.s. of eq.(9), we obtain with necessity the condition  $e^{i2\theta} = -1$ , i.e. the angle between the polarization ellipse major semiaxes should be right ( $\theta = \pi/2$ ). Thus, the general universal bichromatic field configuration for the pure dark state ( $|dark^{(m)}\rangle$ ) generation is  $\varepsilon_1 \perp \varepsilon_2$  configuration. The frequency component amplitudes  $E_{1,2}$

can be arbitrary, and the ellipticity parameters  $\varepsilon_{1,2}$  obey the relationship:

$$\frac{1+F-m}{1+F+m} = \frac{\tan(\varepsilon_1 + \pi/4)}{\tan(\varepsilon_2 + \pi/4)}. \quad (11)$$

Let us distinguish the following particular configurations.

A) The symmetric  $\varepsilon \perp (-\varepsilon)$  configuration, where the polarization ellipses are the same, but with opposite rotations of the field vector. Inserting  $\varepsilon_1 = -\varepsilon_2 = \varepsilon$  in eq.(11), we find the condition:

$$\sin(2\varepsilon) = -m/(1+F), \quad (12)$$

which couples the ellipticity parameter  $\varepsilon$  with the quantum numbers  $m$  and  $F$ .

B) The configuration  $\text{lin} \perp \varepsilon$  ( $\varepsilon \perp \text{lin}$ ), where one of the frequency component is linearly polarized ( $\varepsilon_1 = 0$  or  $\varepsilon_2 = 0$ ). From (11) we obtain:

$$\tan \varepsilon = \pm m/(1+F), \quad (13)$$

where the sign  $(+)/(-)$  corresponds to  $(\text{lin} \perp \varepsilon)/(\varepsilon \perp \text{lin})$ , respectively.

Apart from these, the field configurations  $\sigma_+ - \sigma_+$  or  $\sigma_- - \sigma_-$  (where both frequency components have the same circular polarization, i.e.  $\varepsilon_1 = \varepsilon_2 = \pm\pi/4$ ) obey obviously the condition (11). However, these cases are degenerate, because in addition to the superposition states  $|\text{dark}^{(m)}\rangle$  there exists the trap dark state (the end Zeeman state)  $|F+1, m = \pm(F+1)\rangle$  insensitive to the frequency difference  $(\omega_1 - \omega_2)$ . As a result, one can not prepare a pure superposition state and the atomic state will always correspond to a statistical mixture of  $|\text{dark}^{(m)}\rangle$  and  $|F+1, m = \pm(F+1)\rangle$ .

The two-photon 0–0 resonance is of great importance in view of possible applications to atomic clocks. Let us consider this resonance separately. Inserting  $m = 0$  in eq.(11), we see that  $\varepsilon_1 = \varepsilon_2 = \varepsilon$ , i.e. the general configuration is  $\varepsilon \perp \varepsilon$ , where the frequency components have the same ellipticity and direction of rotation. Particular case of  $\varepsilon = 0$  corresponds to the  $\text{lin} \perp \text{lin}$  field configuration, which has been used in several works [6, 7, 8] in order to increase the dark resonance contrast. For instance, Zanon et al. [8] used pulsed  $\text{lin} \perp \text{lin}$  field for the observation of the two-photon Ramsey-type resonances. The push-pull optical pumping method developed in [7] is also equivalent to the  $\text{lin} \perp \text{lin}$  configuration at the harmonic amplitude modulation. It becomes clear, when the Fourier analysis is applied to the problem. The dark resonance contrast increasing method described in our papers [6] is based on the constructive interference of the two-photon transitions excited by counter-propagating waves with orthogonal circular polarizations ( $\sigma_+ - \sigma_-$  standing wave). This method can be also interpreted in terms of the bichromatic  $\text{lin} \perp \text{lin}$  configuration, which is formed here locally in space, contrary to [7, 8].

It is interesting to note that the 0–0 dark state can be generated by a multi-frequency field with arbitrary

number of equidistant components. The frequency difference has to coincide with the 0–0 transition frequency, the amplitudes have to be the same as well as the phase differences between adjacent harmonics. The frequency component polarizations are chosen in such a way that any pair of adjacent components forms the  $\varepsilon \perp \varepsilon$  configuration. Under the above described conditions the state  $|\text{dark}^{(0)}\rangle$  is dark for any pair of adjacent components as well as for the whole frequency comb. Such a frequency comb can be formed by the push-pull optical pumping method [7] with the pulsed amplitude modulation.

From eq.(11) it follows that the parameters  $\varepsilon_{1,2}$  in the  $\varepsilon_1 \perp \varepsilon_2$  configuration do not depend on the excited state angular momentum  $F_e$ . Nevertheless, the explicit form of the dark state  $|\text{dark}^{(m)}\rangle$  can depend on  $F_e$  in general case. As is seen from eq.(6), this dependence is governed by the ratio of dipole moment matrix elements:

$$\frac{d_{F_1 F_e} C_{F_1 m, 11}^{F_e m+1}}{d_{F_2 F_e} C_{F_2 m, 11}^{F_e m+1}}. \quad (14)$$

Such a dependence (if present) leads to spectacular consequences, if the excited-state hyperfine structure is not spectrally resolved, as it usually is in experiments in the presence of a buffer gas.

Using the well-known formula (see e.g. [12]) for the reduced dipole moment elements ( $d_{F_1 F_e}$  and  $d_{F_2 F_e}$ ), one can see that for the D2 line of alkali metal atoms the ratio (14) does depend on the excited-state angular momentum  $F_e$ , and it equals to

$$-\frac{(F - F_e + 2)(F + F_e + 3)\sqrt{1 + F + m}}{(F - F_e - 1)(F + F_e)\sqrt{1 + F - m}}. \quad (15)$$

Consequently, the dark state  $|\text{dark}^{(m)}\rangle$  will explicitly depend on  $F_e$ . Due to this reason a common dark state, nullifying the interaction with the field via all the excited-state hyperfine levels, is not in existence, as it is illustrated by Fig.2a with the 0–0 resonance in D2 line of  $^{133}\text{Cs}$  as an example. In Fig.2a one can clearly see that the ratio of the dipole moment matrix elements for the  $\Lambda$  system formed via the sublevel  $|F_e = 3, \mu = 1\rangle$  differs from the similar ratio for the other  $\Lambda$  system formed through  $|F_e = 4, \mu = 1\rangle$ . Moreover, in the D2 line there are cycling transitions to the levels  $F_e = F - 1, F + 2$  (see in Fig.2a). According to the selection rules, these transitions can not form the two-photon resonance, rather they destroy additionally the ground-state coherence. Thus, in the absence of the spectral resolution in the excited state the pure superposition state  $|\text{dark}^{(m)}\rangle$  can not be realized in the D2 line.

A completely different type of situation occurs in the D1 line (see in Fig.2b), where the ratio (14) does not depend on  $F_e$ :

$$\frac{d_{F F_e} C_{F m, 11}^{F_e m+1}}{d_{(F+1) F_e} C_{(F+1) m, 11}^{F_e m+1}} = -\sqrt{\frac{1 + F + m}{1 + F - m}}. \quad (16)$$

For example, the dark state  $|dark^{(m)}\rangle$  in the symmetric  $\varepsilon \perp (-\varepsilon)$  configuration (12) for any  $m$  has the form:

$$|dark^{(m)}\rangle = N\{|F, m\rangle - i(E_1/E_2)|F+1, m\rangle\}. \quad (17)$$

Note, the substate populations are controlled by the frequency component amplitudes  $E_1$  and  $E_2$ . Thus, in the D1 line the dark superposition states  $|dark^{(m)}\rangle$  can be prepared even in the absence of the spectral resolution in the excited state. What is a key advantage of the D1 line (with respect to the D2 line) in the dark resonance observation.

Besides, the dark state  $|dark^{(0)}\rangle$  with  $m = 0$  in the D1 line can be generated even if the ground-state hyperfine structure are not spectrally resolved [7]. To do this one has to use the above described  $\varepsilon \perp \varepsilon$  frequency comb with the total spectral width much larger than the optical linewidth  $\Gamma_{opt}$ . For instance, in the method of the paper [7] it corresponds to the amplitude modulation by pulses of duration  $\tau \ll 1/\Gamma_{opt}$ .

#### IV. OTHER POSSIBLE VARIANTS

For the sake of completeness, we give here the results for other possible variants of the ground-state momentum values  $F_1$  and  $F_2$ , which are allowed by the selection rules for the two-photon resonance of  $\Lambda$  type. These results are follows from the analysis of eq.(9). In practice such variants can be realized, in principle, in several elements with complicated ground-state structure, and also with the use of metastable levels.

The selection rules allow the following combination of the total angular momenta:  $F_1 = F$ ,  $F_2 = F + 2$ ,  $F_e = F + 1$ . In this case the dark state  $|dark^{(m)}\rangle$  is generated by the  $\varepsilon_1 || \varepsilon_2$  field configuration, where the major ellipses semiaxes are parallel ( $\theta = 0$ ), and the ellipticity

parameters  $\varepsilon_{1,2}$  obey the condition

$$\frac{(1+F-m)(2+F-m)}{(1+F+m)(2+F+m)} = \frac{\tan(\varepsilon_1 + \pi/4)}{\tan(\varepsilon_2 + \pi/4)}. \quad (18)$$

The amplitudes  $E_{1,2}$  can be arbitrary.

The last variant is  $F_1 = F_2 = F$ , when the two-photon resonance can be excited via the excited-state level with different momenta  $F_e = F, F \pm 1$ . Here for any  $F_e$  the dark states are generated by the common (independent of  $m$ ) filed configuration  $\varepsilon || \varepsilon$ , where the frequency components have the same elliptical polarization.

#### V. CONCLUSION

We have proposed the bichromatic field configurations, allowing to prepare (with the use of the CPT effect) the pure superposition  $m-m$  states for arbitrary magnetic quantum number  $m$ . It has been shown that in the general case of  $m \neq 0$  it is necessary to use elliptically polarized fields. It has been found that for alkali metal atoms in the absence of the excited-state spectral resolution the superposition dark states can be only generated in the D1 line. The obtained results have a wide spectrum of applications from atomic clocks and magnetometers to quantum informatics. The preparation of the pure  $m-m$  states looks especially attractive in magnetometers, where it allows one to get a high-contrast resonance with maximal sensitivity to a magnetic field.

Recently, we have got first experimental evidence of the described theoretical predictions. The corresponding results will published elsewhere.

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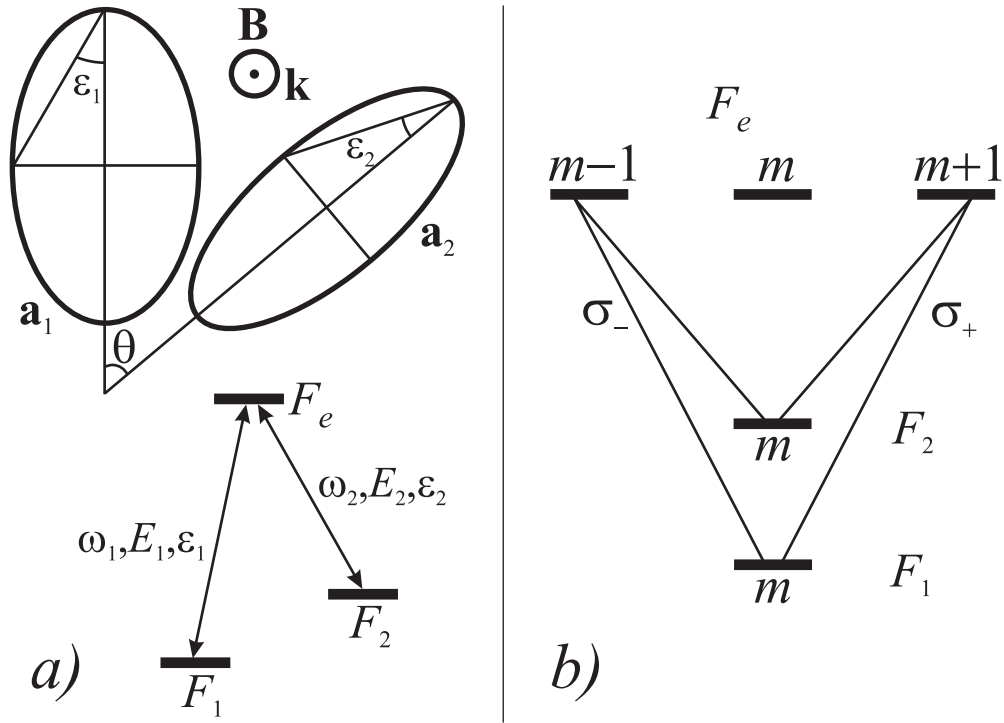


FIG. 1: Illustration to the problem statement: *a*) mutual orientation and parameters of the polarization ellipses ( $\mathbf{a}_1, \mathbf{a}_2$ ), and the general scheme of optical transitions, exciting the two-photon resonance of  $\Lambda$  type; *b*) scheme of the light-induced transitions driven by the  $\sigma_+$  and  $\sigma_-$  circularly polarized components for the  $m-m$  resonance.

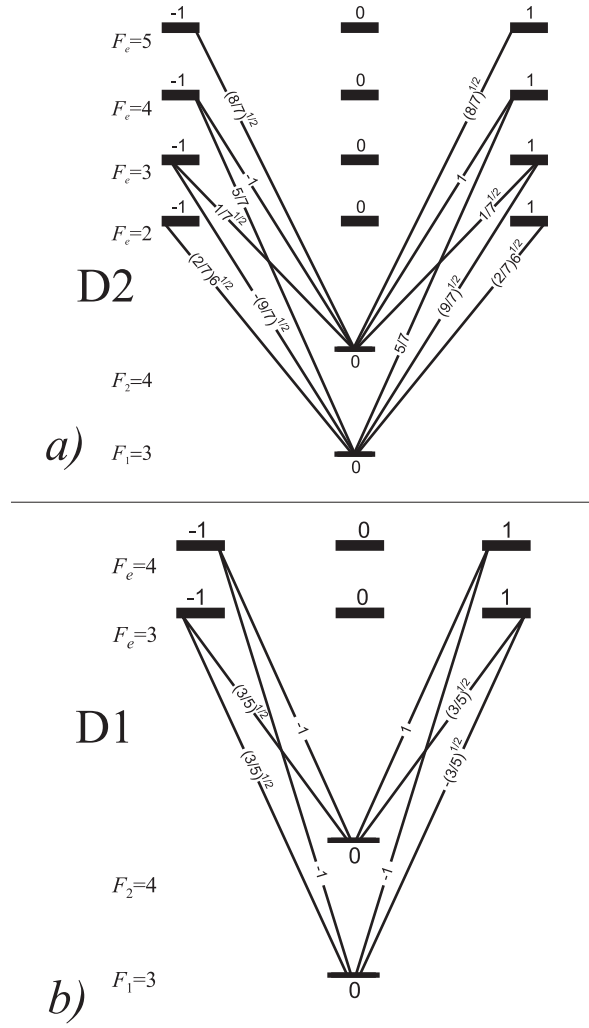


FIG. 2: Scheme of the light-induced transitions from the states  $|F_1, m=0\rangle$  and  $|F_2, m=0\rangle$  in bichromatic field. Numbers on lines denote the relative values of the dipole moment matrix elements for  $^{133}\text{Cs}$ : a) in the D2 line; b) in the D1 line.